

WEIGHTLESS, CONTAINERLESS MELTING AND SOLIDIFICATION OF POTENTIAL NEW METAL AND CERAMIC PRODUCTS

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ABSTRACT

A number of suggestions have been made for the weightless processing in space of metals and ceramics. The most obvious application of a weightless environment is the removal of buoyancy forces in multi-phase systems and the possibility for containerless melting and solidification processes. Under the first category, processes which can be considered are the casting of superalloy materials containing rare earth oxide inclusions and the casting of metal emulsions where convection currently leads to phase segregation. Under containerless processing we can consider extensions and scale up of processes which have been studied in terrestrial levitation experiments. Possible applications are the preparation of ultrapure materials and solidification under conditions of extreme subcooling. Ultrapure refractory materials which are easily contaminated by molds are logical candidates. Control of grain size by solidification at extreme subcooling is another possibility if a means of inhibiting subsequent grain growth can be devised. Preparation of high density glasses and ceramics by containerless solidification can also be considered.

Initial results in exploring the physical problems associated with weightless, containerless processing in the space environment are described. Analyses and experiments are described which treat the possibilities and limitations for eddy current positioning and handling, surface oscillations and fluid currents affecting the motion of bubbles, scale up and power requirements as a function of material properties such as density, surface tension, viscosity, electrical conductivity, dielectric constant and permeability.

INTRODUCTION

A number of suggestions have been made for the exploitation of the weightless feature of the space environment to develop new or improved processes for preparation of materials. A number of these suggestions involving the melting and solidification of a free floating mass of metal or ceramic involve the handling or transfer of such a mass during some stage of the process. In addition to a more general study of feasible materials preparation processes at General Electric we have begun to study in some depth the initial experiment techniques hardware which will be required to exploit these ideas in space. In order to be able to provide sufficient depth in our study we have at first limited ourselves to the study of the physical phenomena and required experiment hardware associated with the free floating phase of a crucibleless experiment.

Figure 1 lists four general areas of phenomena which can be considered which exploit the weightless state. The most obvious application is the elimination of buoyant segregation in multiphase systems where present forming techniques are limited by separation of phases of different densities. Materials of this type often must be prepared by powder metallurgy techniques which result in low ductility and creep resistance or which prevent molding into precision shapes.

The second example refers to the new possibility of carrying out precision molding of reactive and high melting materials which presently must be prepared by skull melting techniques which do not allow for the provision of superheat. This severely limits the shapes which can be cast. In a free floating state superheat could be provided even for very reactive materials.

A more speculative possibility is the achieving of high degrees of subcooling prior to solidification because of absence of contact between the melt and any solid object. High degrees of subcooling have been observed in limited

terrestrial levitation experiments and new crystal phases have been observed such as Gallium III, to be formed at subcooled temperatures. Another possibility which has been suggested is the preparation of high index of refraction glass where recrystallization due to mold contact is a present limitation. I will show a few specific suggestions in the first two categories by way of introduction to a discussion of the physics and hardware which must be provided for first experiments in these areas.

Figure 2 lists some examples which have been suggested for exploitation of the removal of buoyant segregation of multiphase materials. There is currently some interest in this possibility on the part of our aircraft turbine people and some recent studies have been devoted to this at our Research Laboratory. Accelerated corrosion in a Marine environment can severely limit jet engine life because of blade and vane parts which are of thin walled cast construction and because of the high operating temperatures. Wasielewski has studied the oxidation and hot corrosion of superalloys and has documented the beneficial effects which the rare earth metals have on the hot corrosion process. Additions of Lanthanum and Cerium as metals, however, lead to low melting eutectics at grain boundaries, severely limiting the high temperature capabilities of the alloy. Through these initial efforts, it had been recognized that the rare earth oxides are the effective barrier rather than the metal itself. Work by Seybolt and Allen has further delineated these effects and has shown that the protection mechanism depends wholly on the presence of oxide and that the dispersion or size of the particle need not be critical. Accelerated corrosion occurs as a result of combining sodium with the sulfur in fuels to form sulphates which then rapidly deplete chromium from the matrix as shown in Figure 3. Sulfur from the fuel in the presence of salt diffuses into the matrix and depletes the chromium from the matrix leaving it in the form of chromium sulphate. When the oxide dispersion is added the sulfur is taken up by the cerium and the oxygen is released to oxidize the chromium. The chromium oxide is far less deleterious to the metal matrix than is the chromium sulfide.

Presently, because of the large difference in density between rare earth oxides and superalloy mixtures, only powder metallurgy techniques can be utilized to form oxide-metal dispersions. Figure 4 is a dramatic demonstration of the oxidation/hot corrosion problem and the protective role which the rare earth oxides play. These two specimens of TRW-6A alloys (a modern superalloy developed by Thomson, Ramo, Woolridge) were fabricated from prealloyed powders to one of which 1/2% by weight CeO_2 was added. These samples were initially 1/4" diameter rods which were exposed to 1725° for fifty hours with 100 ppm NaCl added to the intake gases. Present conservative estimates are that rare earth additions to superalloys will more than double their useful life.

Casting of the material including the rare earth oxide leads to segregation due to buoyancy effects. Weightless solidification of such a material can thus be considered as a potential interesting process if means can be found to make castings in a nearly zero gravity environment.

The casting of other materials in a nearly weightless environment in order to improve properties and materials presently limited by powder metallurgy techniques can also be considered. Examples might be nickel thoria and aluminum-alumina materials.

Another general area for consideration of weightless metallurgical processing is the crucibleless melting reaction of high melting point metals. Figure 5 gives some examples. Present skull melting techniques without the possibility of providing superheat generally exclude precision casting. One metal which is currently of great interest to General Electric for switching and conductor applications is pure beryllium. Present high voltage underground distribution systems in cities are severely limited by the ohmic resistance of aluminum and copper and our company has recently carried out a study for the Edison Electric Institute on the possibility for cryogenically cooled transmission lines which would reduce the joule heat by two decades or so. Figure 6 shows qualitatively the variation of electrical resistivity of copper, aluminum and beryllium as a function of temperature. Cooling at liquid hydrogen temperatures would be necessary in order to reduce the resistivity of aluminum or copper by the required amount. For beryllium on the other hand sufficiently low resistivity for desired applications can be achieved at liquid nitrogen temperatures which appear much more practical. The possibility for the crucibleless formation of a billet of this material free from contamination and in a form suitable for later extrusion seems to us worthy of further study.

Many optical devices would benefit from the use of isotropic transparent materials (glasses) with high index of refraction. Thin lenses would be one feature and would lead to smaller optical instruments. Many glassy systems are known to have high index of refraction but they are usually susceptible to devitrification (recrystallization). Most recrystallization in glassy materials is surface nucleated from contact with crucibles in which melts are made. Levitation melting offers the ability to melt out of contact with crucibles and to explore the possibility of glass forming from compositions which normally would be considered unstable. The possibility also exists that molten materials, which do not normally form amorphous, glassy solids, would do so if they were supercooled and solidified out of contact with crucible materials.

Examples of compositions that are worthy of study in this context are $\text{PbO}:\text{SiO}_2$ systems rich in PbO (max. RI=2.6) and $\text{PbO}:\text{TeO}_2$ systems. Glasses of RI=2.2 have been obtained from these systems by normal glassforming techniques.

ELECTROMAGNETIC POSITIONING OF A FREELY FLOATING MASS

The basic theory associated with the application of forces to a conducting sphere by eddy currents has been treated by Smythe in his textbook on electromagnetic theory. Figure 7 shows a spherical object, here considered to be a conducting sphere located between circular coils excited with audio or radio frequency currents. I shall later say a few words about position control schemes for dielectrics such as glass or ceramics. Smythe's treatment assumes an oscillating magnetic field in the direction of the coil axis uniform over the sphere volume. The analysis is best done in terms of the magnetic potential A rather than the magnetic field B . The vector potential A has a circular configuration centered about the coil axis and is considerably simpler to compute, given the coil configuration, than is the magnetic field itself. The force per unit volume within the conducting mass is given by the vector product of current density and magnetic induction B . From electromagnetic theory we can write the electric current density as $\vec{j} = \frac{\sigma}{c} \frac{\partial \vec{A}}{\partial t}$ where σ is the

conductivity. The magnetic induction B is given by $\vec{B} = \text{curl } \vec{A}$. The total force acting on the sphere can be obtained by integrating the force per unit volume over the sphere. There is also a simpler method. However, we are interested in the force per unit volume since its distribution is important in setting up fluid current circulation patterns within the molten mass. The body distributed forces will affect the motion of bubbles which may be included in the melt and bubble motion from this will probably be more important than bubble motion due to rotation of the mass or other effects. The nonuniform distribution of forces can also give rise to shape oscillations of the spherical mass which I shall discuss later.

We have developed a servo control which applies translational forces to the sphere based on sensing of position errors by means of the mutual induction between the eddy currents within the sphere and the driving coils. Since the electric field is simply the time derivative of A it will be in the same direction as A and hence the current will flow in circular loops of the type shown for A on the slide. The summation of these circular currents can be thought of as a single winding transformer secondary which will have a reflected impedance as viewed from the driving coil terminals. The power dissipation per unit volume W is given by the resistivity (reciprocal sigma) times the square of the current density.

We have plotted the average current distribution (Figure 8) for a radial traverse in the equatorial plane. The curve labeled (1) is the cycle average distribution within a .4 inch radius aluminum ball when the frequency is 11 kilocycles. The current waves propagate along the radius and die out rapidly

a short distance within the spherical boundary. The length within which the waves attenuate to $1/e$ of their surface value is called the skin depth and for this particular curve the ratio of sphere radius to skin depth is 13. At lower frequencies the skin depth increases and the current remains flat as we proceed inward. At lower audio frequencies the magnetic field is nearly uniform throughout the sphere.

We have also plotted the distribution of instantaneous magnetic field within the sphere. At the lower frequencies the field decreases only slightly within the sphere. At the higher frequencies the field reverses as we travel into the sphere. (Figure 9)

We have also computed the vector product of the electric current and magnetic field within the sphere. We find that the resultant force field within the sphere is rather complex at the higher frequencies. I shall discuss this later. At the lower frequencies the force distribution within the sphere becomes simple. The force is radially inward at the surface of the sphere. We are dealing with the phenomenon of magnetostriction which will give rise to fluid currents within the sphere, spheroidal shape oscillations and sound wave generation. It is only for the latter phenomenon that we are interested in the instantaneous forces as they vary at audio or radio rates. For prediction of fluid currents, and excitation of low frequency shape oscillations, we are more interested in the average forces over a full cycle of oscillation of the applied field. We have also computed these cycle averages for some interesting cases.

Because of the necessity of controlling the position of the processed material in relation to a heat source or other apparatus and to compensate for small accelerations due to spacecraft station keeping, attitude motions, astronaut motions, or location other than at the vehicle center of mass, it will be necessary to provide position sensing and control devices. We have developed a position sensing system based on electromagnetic principles so it is possible to position sense and control even without optical windows. This might be highly desirable when processing materials which will outgas and tend to plate optical windows. The force exerted by a control coil on a nonmagnetic conducting sphere depends of course on the coil current, dimensions, frequency, sphere radius and sphere conductivity. The frequency and conductivity enter the problem in determining the skin depth of penetration of electromagnetic waves into the sphere. Figure 10 shows theoretical curves for the total force on an aluminum sphere with the parameter R_2/δ which gives the ratio of sphere radius to skin depth and where the abscissa is the ratio of coil to sphere separation divided by coil radius. The ordinate has been expressed in terms of acceleration of an aluminum sphere per unit ampere turn of coil excitation.

For position control in a weightless environment we can scale very low values of control acceleration--for example, in our initial tests which I will describe we use an ampere turn value on the order of several hundred which gives an ampere turn squared value of about 10^5 . This means that the maximum ordinates 10^{-3} cm sec $^{-2}$ would be multiplied by 10^5 and this level on the ordinant scale would become 100 cm sec $^{-2}$ or about 1/10g. Values of a few tenths g are easily obtained with a few watts of control power at higher frequencies which give a small skin depth. Since for most applications we can be satisfied with accelerations below 10^{-3} or 10^{-4} g, these curves illustrate that we can consider the use of very low audio frequencies which give skin depths equal to the sphere radius or greater.

Position sensing makes use of the relative phase change between voltage and current in the driving coil due to variation in the reflected impedance from the eddy current loops as the ball position changes. Figure 11 shows the equivalent circuit of the driving coil which is resonated by the use of a tuning capacitor. Of course as the ball position varies exact resonance is not maintained. This curve shows the results of measurement of the sensitivity for position detection by a single driving coil. We see that displacements of a millimeter give a phase change on the order of a degree or so. In our control servo scheme it is this phase change that is detected.

The amplitude of current and voltage in the driving coil depend not only on the ball position but also on the excitation level of the coil. By using phase changes as a position indicator we free ourselves from the dependence on the level of coil excitation and allow for the possibility of using the same coils for RF heating of the sample, if desired.

We have made a number of force measurements for aluminum and copper spheres at various frequencies and coil sphere distances to compare with theory. Figure 12 shows forces measured by a balance technique for an aluminum sphere and frequencies between 10 kHz and 100 Hz. The sphere to coil separations are given in dimensionless form by dividing by coil radius. The force is normalized to 1 ampere turn although in these experiments ampere turn values up to several hundred were used. Shown as the dashed line is a theoretical calculation based on a computation assuming that the currents within the sphere are excited by a uniform oscillating magnetic field. There is some inconsistency in computing the net translational force for the case where the applied field is uniform and this partially explains the discrepancy shown here. Actually if the applied field were uniform there would be no net translational force on the sphere. The procedure which is used in the literature is to compute the currents in the sphere for the uniform field case but then to assume that these currents are acted upon by the nonuniform field near the coil so that a net force is observed.

Figure 13 shows experimental data in a smaller force regime. We have extended the ability to measure forces down to the 10^{-4} to 10^{-5} g regime since for many applications control accelerations of this magnitude will suffice and we wish to size space experiment hardware for these realistic cases. To this end we have begun measurements using a ball suspended as a long pendulum for measurement of weak horizontal forces.

Figure 14 shows the pendulum set up. Here we see some positioning coils, the pendulum enclosed by a plastic case to eliminate the effect of air currents and a telescope for measuring small position displacements. An even more sensitive technique for measuring small horizontal forces is the use of very perfect spheres on the glass tilt table whose tilt is adjusted by 3 micrometer screws serving as legs. Although the measurement of such small forces can be avoided for positioning studies of conducting spheres, for poor conductors and dielectrics such as glass it is easier to measure the small forces rather than scaling up the fields. The coils shown in this set up have been used only for the purpose of observing the operation of the position servo which I will now describe. The actual coils used for metallurgical space processing experiments might be water cooled and fed by transformers to match them to the driving source.

In the zero gravity environment, position sensing and control of a floating mass before and after melting and solidification would be done by three orthogonal sets of coils. If all three sets of coils are excited about equally, the floating mass will find itself in a position of stable equilibrium near the center of the coil configuration. The damping time for positioning oscillations, however, tends to be rather long so that we have designed a servo device which can correct position errors with any desired damping constant.

Figure 15 shows a block diagram for a single axis control with the two coaxial driving coils on either side of the experiment specimen represented schematically. The phase shift detection of position changes is determined by a logic circuit which compares the phases of driving coil current and voltage. To obtain more accurate position sensing a differential circuit is employed which compares the phase differential detected by the two coils. Depending on the size of the position error the power to one or the other driving coils is increased by a modulator under control of the position error signal.

Figure 16 shows the hardware implementation of this circuitry which is rather standard except for the logic circuitry which is unique to this application. I have already shown measurements of the position sensing characteristics for this circuitry and the position control stiffness in terms of force per unit current. The characteristic resonant frequency of this servo loop can be adjusted if desired by furnishing equal excitation to both coils in the absence of an error signal.

BASIC HYDRODYNAMICS

Oscillations

Excitation of the position control coils can be used either to initiate or damp translational motions of the floating mass and shape oscillations as well as any rotations which may occur. The position control and shape oscillation problem is most easily treated by considering individual hydrodynamic modes of a liquid sphere under the action of surface tension forces and the electromagnetic forces due to internal eddy currents.

Figure 17 illustrates the first three hydrodynamic modes. We can describe these modes most easily by expressing the distance from the center of mass to the liquid surface r as a function of a colatitude θ in the way shown at the bottom of the figure. For the zero mode, the radius vector equals a constant a_0 multiplied by the 0 order Legendre polynomial which is simply unity. For this mode the mass is a perfect sphere with no distortion motions. For the second hydrodynamic mode which represents a shape oscillation between prolate and oblate spheroid, we must add to the original radius a term $a_2 P_2(\cos \theta) \cos w_2 t$. The center figure illustrates this shape at a given instant of time. The shape oscillates sinusoidally between oblate and prolate. This oscillation mode can be either excited or damped by application of the magnetostriction forces thru excitation of the position control coils. In my paper given to this Symposium last year I discussed the limitations arising from excitation of oscillations and showed that we can consider processing of liquid metal masses at least as large as tens of kilograms without worrying about rupture of the mass due to the necessity of position control in practical cases.

If the position control coils along a given axis are excited asymmetrically, for example if we excite only one, then we will obtain a net translational force which will excite both the first and third mode. The third mode resembles an eggplant and can be treated similarly to the second mode. The first mode illustrated to the left represents pure translation of a spherical shape and must be treated differently from the other modes. The reason for this is that the frequency corresponding to the first mode is zero and for this particular term in the sum we should replace the sinusoidal time term by a term linear in time. We have begun to examine theoretically the excitation or damping of these various modes by excitation of the coils.

Internal Fluid Currents

In addition to generation or damping of surface oscillations and translational motions, we can stimulate circulating currents within the floating mass by the magnetostrictive forces which I discussed earlier. Figure 18 shows a simple current pattern which can be stimulated by exciting a single control axis. These fluid currents can be oriented in various directions by exciting various sets of coils. These currents will be useful for stirring of alloys or dispersions and will also affect bubble motion in an important way.

At the higher frequencies the pressure gradient resulting from the magnetostrictive forces which drive the steady state fluid currents will be concentrated in a thin surface layer centered about the equatorial plane with respect to the excitation axis. After the coils are excited an equilibrium will quickly be reached in which this driving pressure gradient is balanced by viscous forces. At lower frequencies where the skin depth becomes an appreciable fraction of the sphere radius the flow patterns will be altered somewhat. The quasi-equilibrium shape of this spheroid will be influenced not only by the magnetostrictive forces which drive the fluid currents but also by the dynamics of the circulating dense fluid. Since magnetostrictive forces can be applied in an arbitrary manner, and translational acceleration of the mass can be suppressed by simultaneous excitation of opposite members in coil pairs, we see that considerable freedom exists for electromagnetic shaping of the floating mass in various symmetric or asymmetric configurations.

Rotation

In the previous paper the excitation or damping of rotations by the phased excitation of coil pairs was discussed.

If we consider an orthogonal pair of excitation coils to be excited with a phase quadrature so as to produce a rotating magnetic field, we can calculate that the spheroid will be caused to rotate in the manner of the rotor of an induction motor. By proper choice of excitation for various pairs of the positioning coils, angular velocity can be imparted the floating spheroid in any desired direction and with any speed consistent with configurational stability of the mass. This will be limited by the stability furnished by the action of surface tension. Oscillational instability can occur for higher rotational speeds and damping due to viscosity will play a key role in prevention of buildup of catastrophic oscillations. Conversely, properly phased excitation of a given pair of coils can be used to remove an angular velocity orthogonal to both coil axes if such rotation is not desired.

Sound Waves

The high frequency variation of magnetostriction forces can be used to generate sound waves within the floating mass if we desire to go to high level excitation. This might be useful, for example, to prevent agglomeration of oxide dispersion particles within the mass as suggested by our research laboratory people.

**POTENTIAL CRUCIBLE-LESS METALLURGICAL/CERAMIC PROCESSES FOR
ZERO-GRAVITY APPLICATIONS**

ELIMINATION OF BUOYANT SEGREGATION

PRECISION MOLDING OF REACTIVE AND HIGH MELTING METALS

**FORMATION OF NEW PHASE OR MICRO-CRYSTAL STRUCTURE THRU
SUB-COOLED SOLIDIFICATION**

PREVENTION OF CRYSTALLIZATION BY AVOIDANCE OF MOLD CONTACT

Figure 1

EXAMPLES OF ZERO-BOUYANCY PROCESSING

- **OXIDE DISPERSIONS IN SUPER ALLOYS**
Ce O₂ in TRW-6A
- **MULTI-PHASE THERMO ELECTRIC ALLOYS**
Pb Sn Te
- **METAL EMULSIONS FOR ELECTRICAL CONTACT APPLICATIONS**
Ag + WC, W etc.
- **DISPERSION HARDENED ALLOYS**
Ni + Th O₂ Al + Al₂O
- **REACTOR COMPONENTS**
Pu O₂ - Mg O

Figure 2

RARE EARTH OXIDE DISPERSIONS IN SUPER ALLOYS

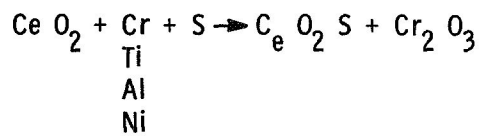
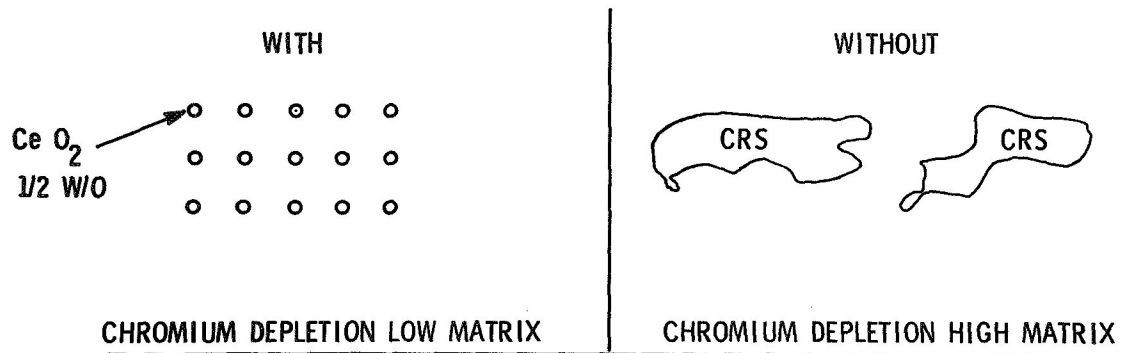


Figure 3

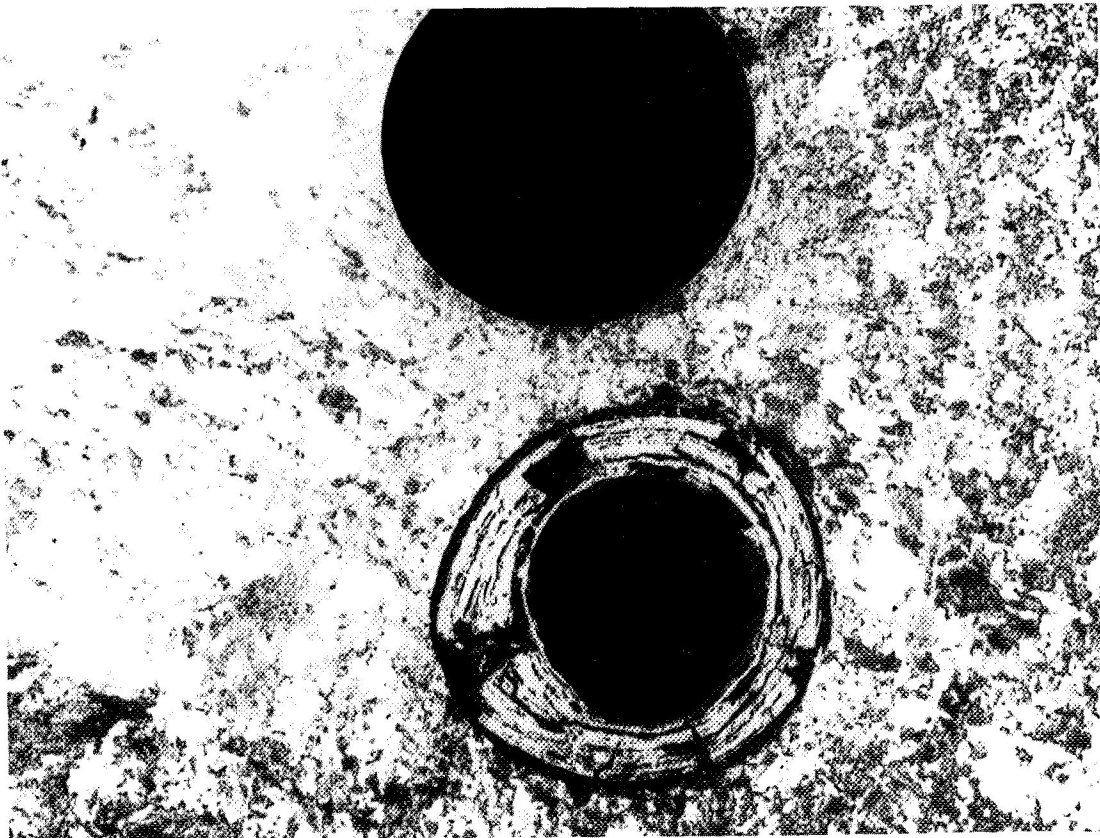


Figure 4

CRUCIBLE-LESS MELTING - PRECISION CASTING, EXTRUSION SHAPES

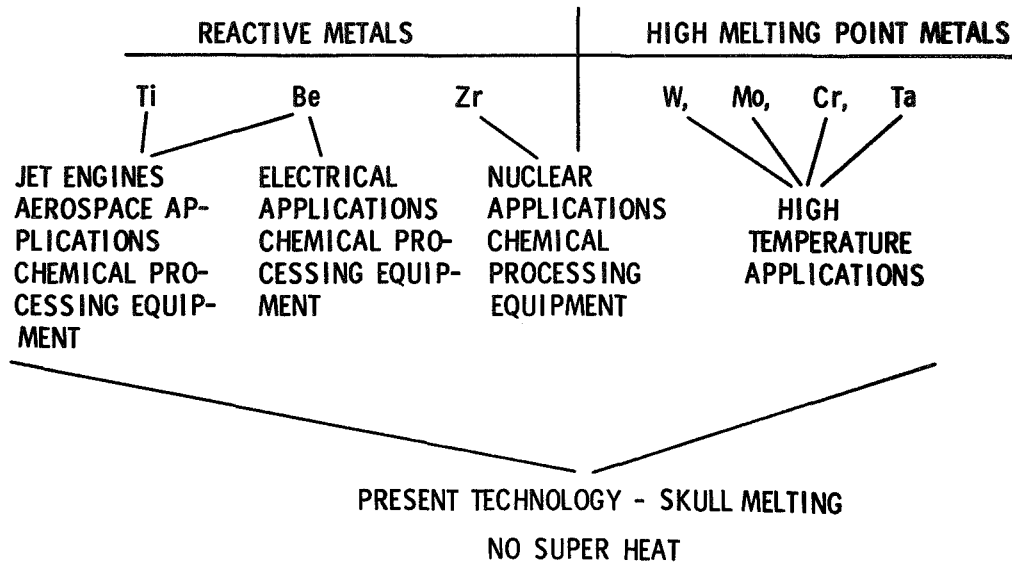


Figure 5

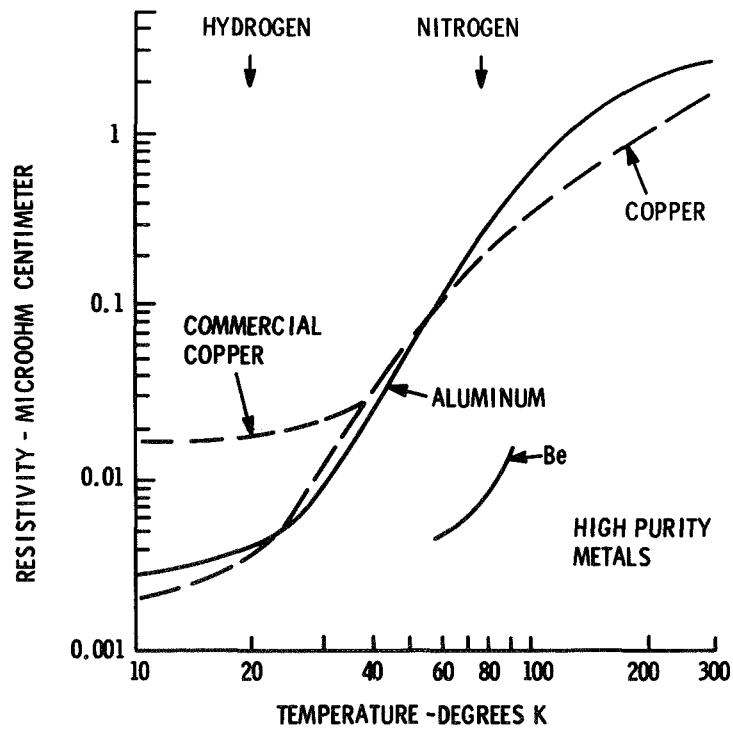


Figure 6

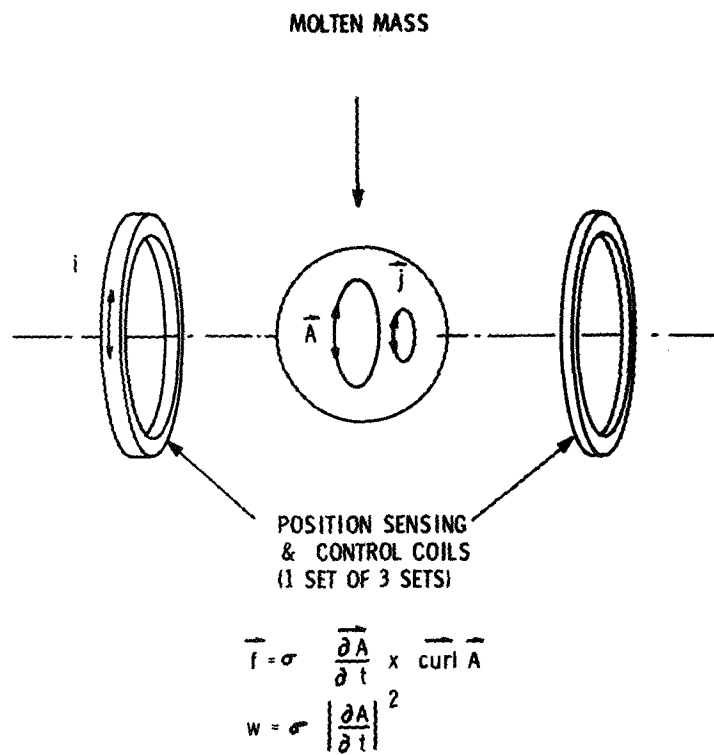


Figure 7

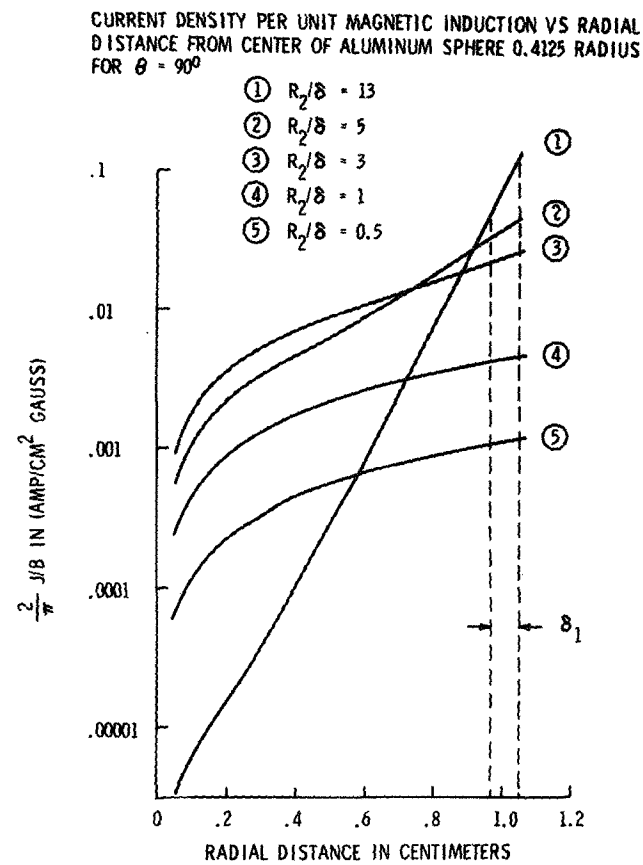


Figure 8

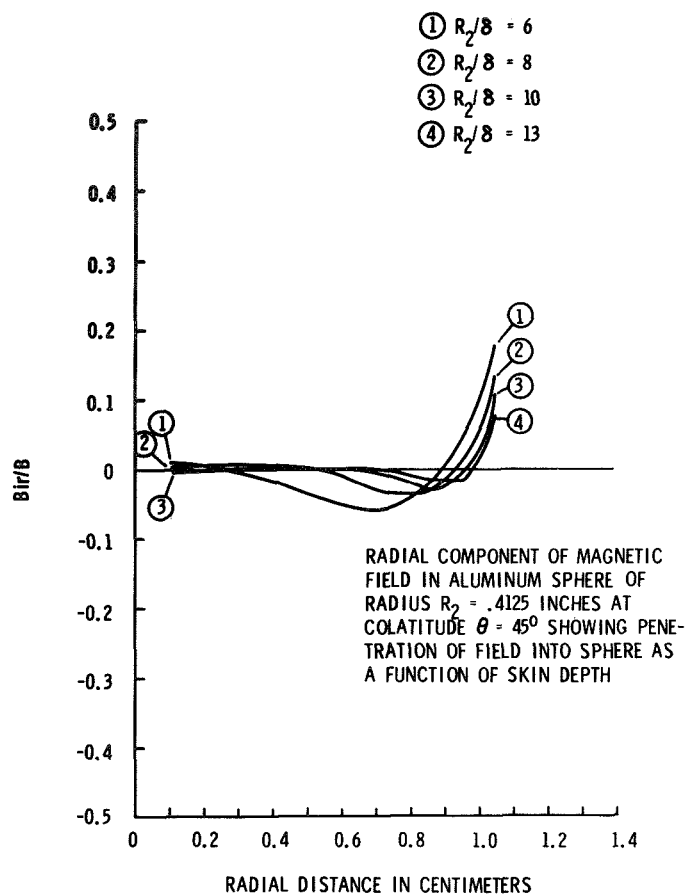


Figure 9

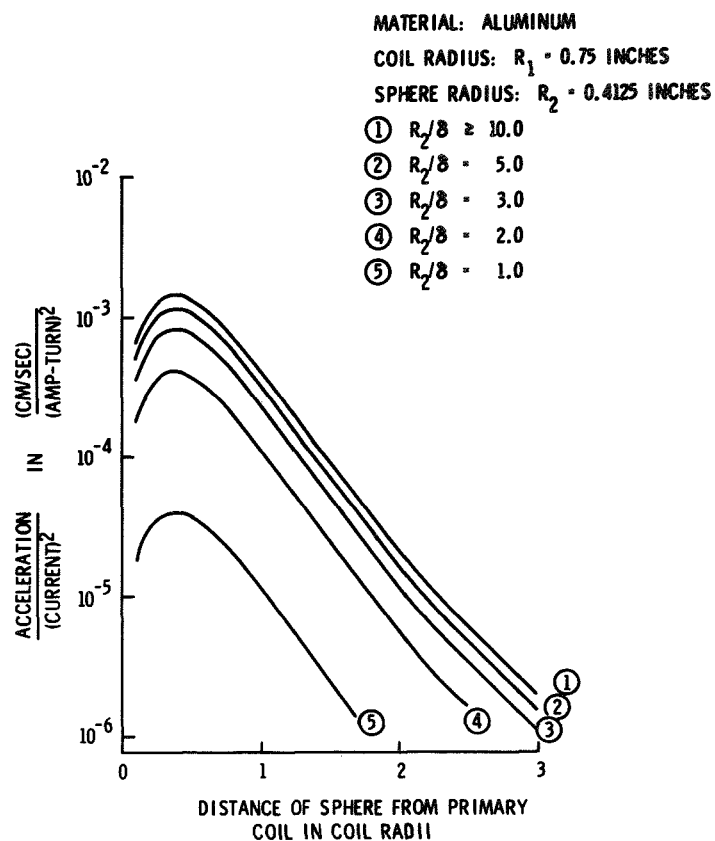


Figure 10

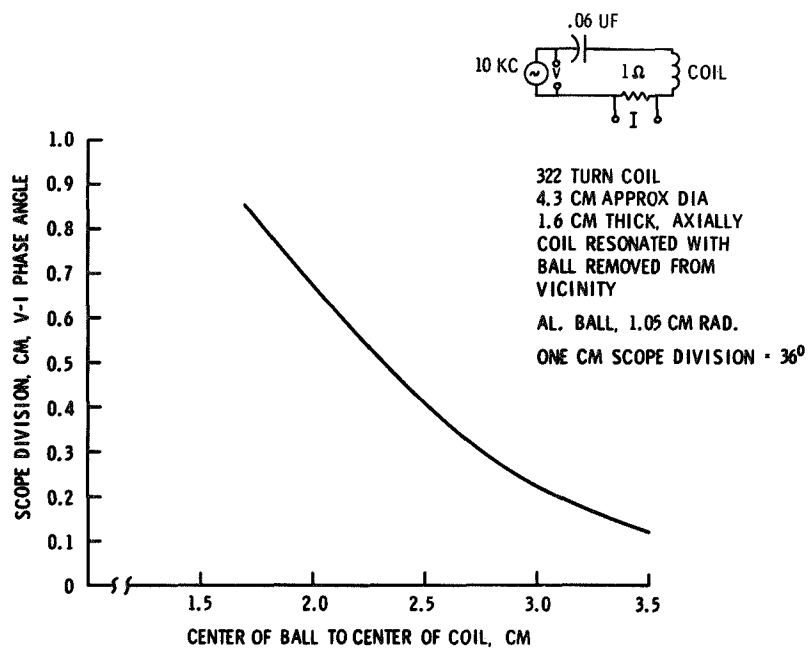


Figure 11

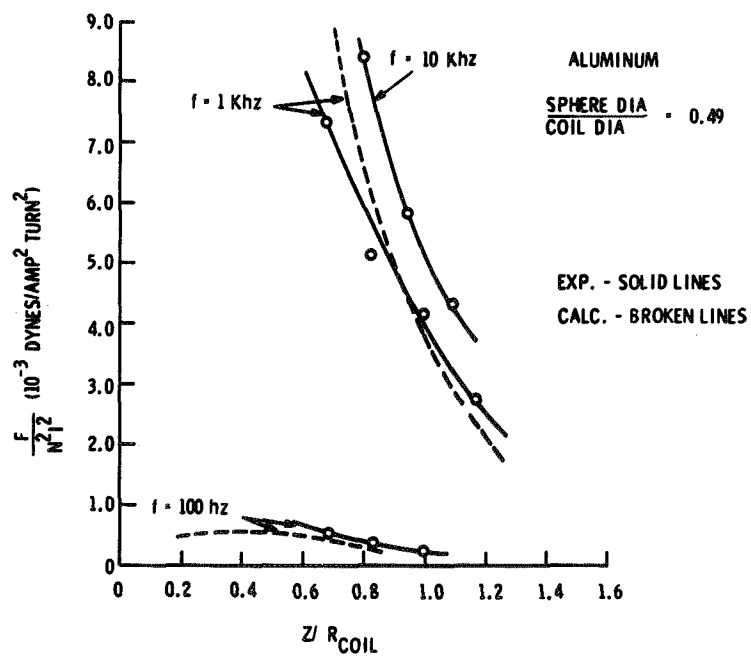


Figure 12

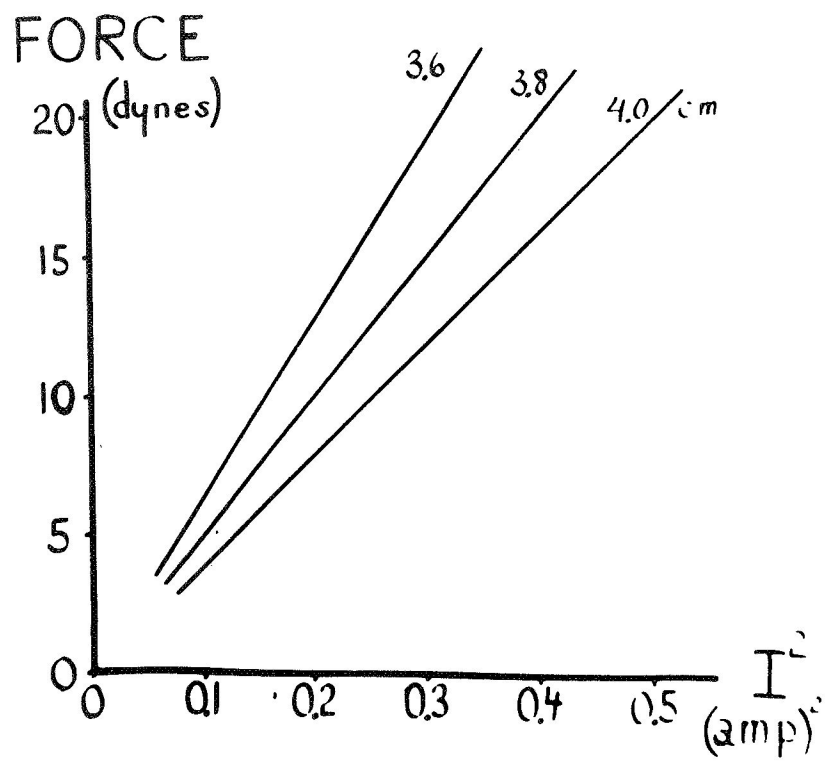


Figure 13

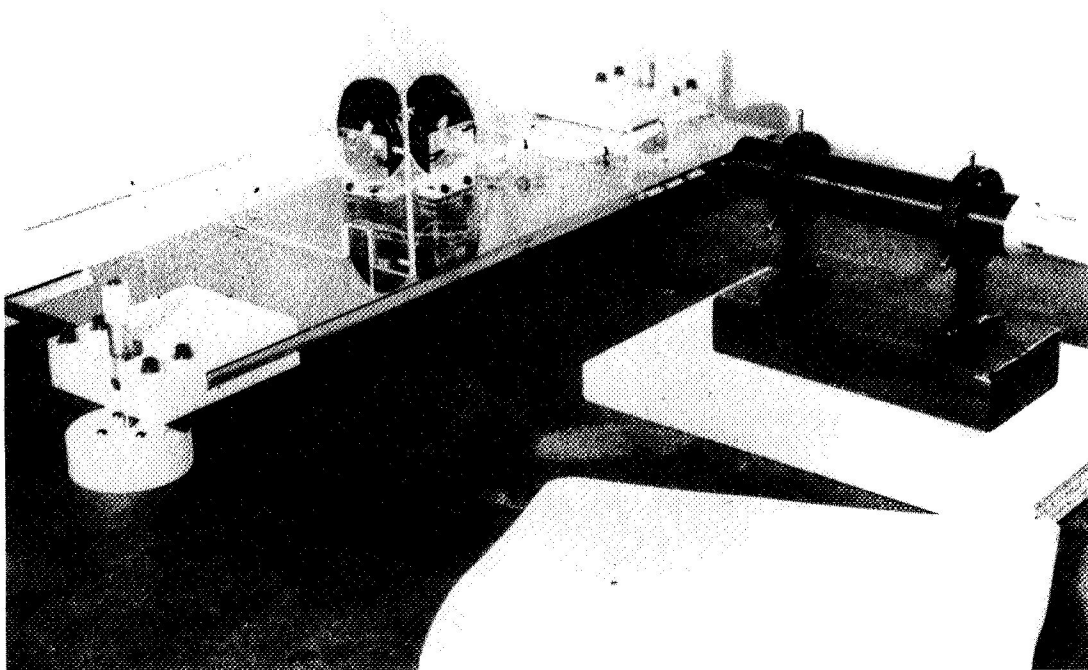


Figure 14

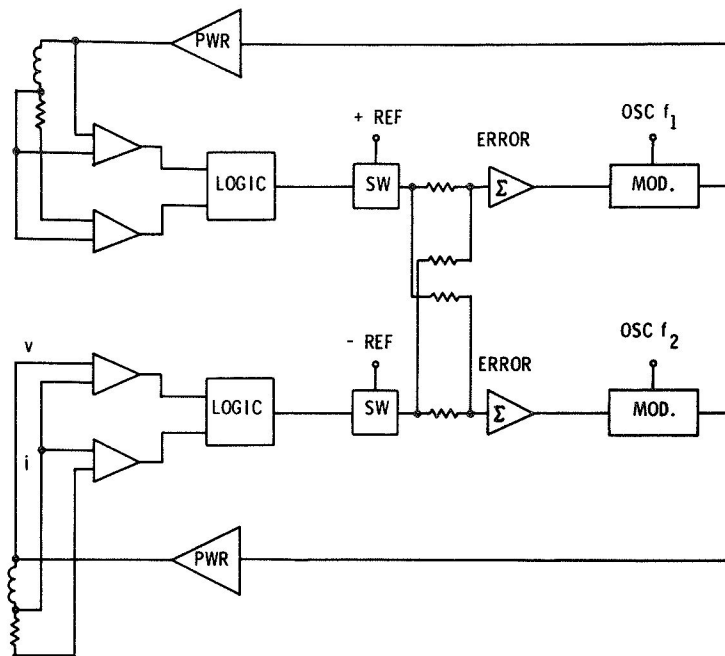


Figure 15

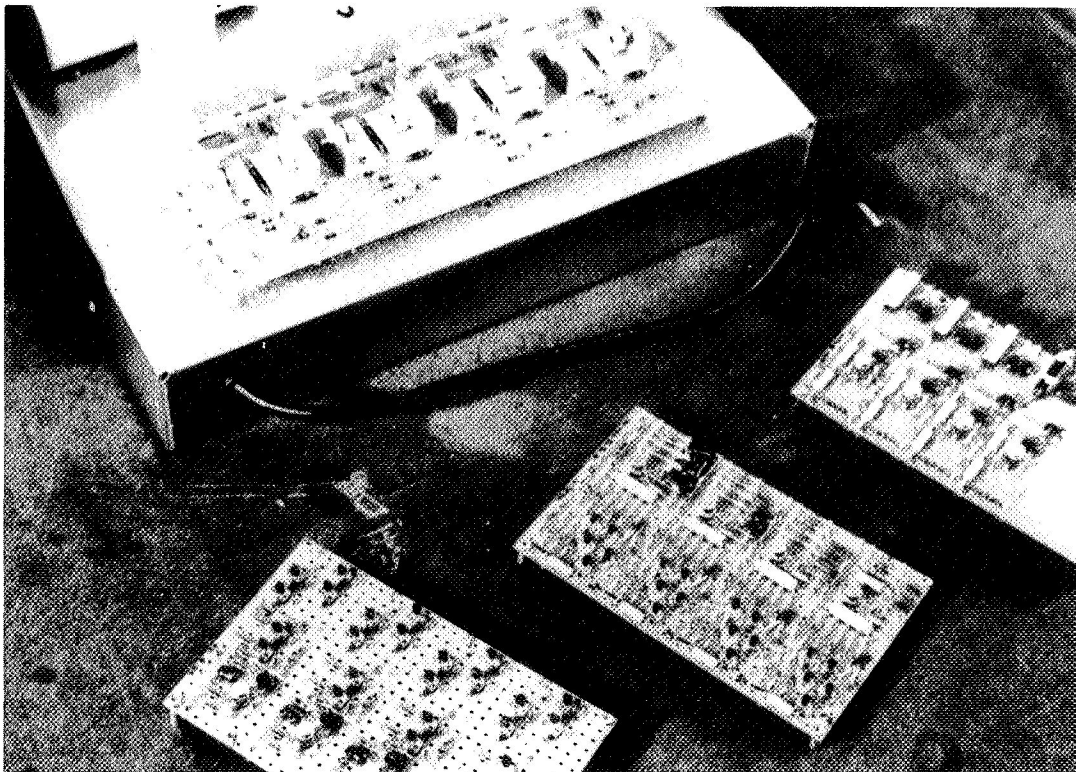
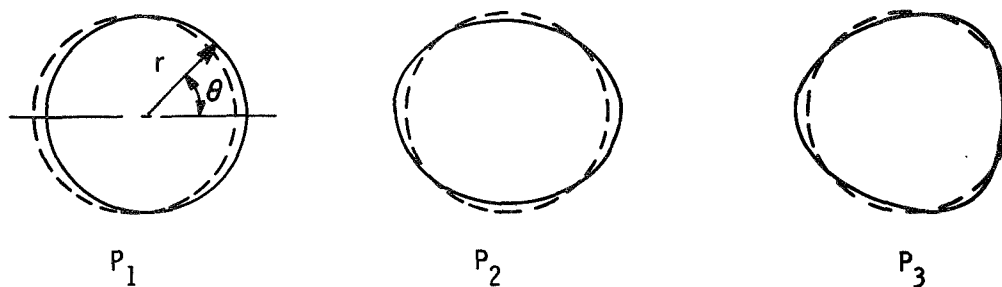


Figure 16

HYDRODYNAMIC MODES



$$r = a P_{\ell}(\cos \theta) \cos \omega_{\ell} t$$

Figure 17

FLUID CURRENTS DUE TO MAGNETOSTRICTION

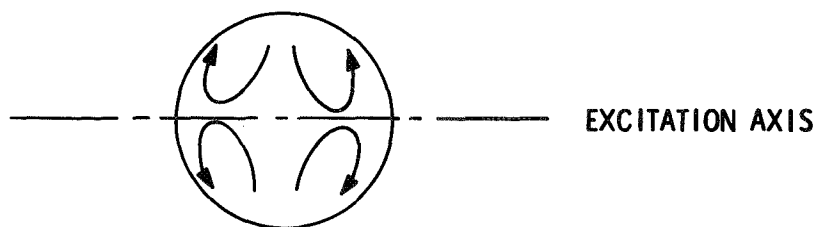


Figure 18